

An Overall Understanding of Sodium Storage Behaviors in Hard Carbons by an “Adsorption■Intercalation/Filling” Hybrid Mechanism

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Abstract

Hard carbon has the potential to serve as a high■capacity anode material for sodium■ion batteries (SIBs), however, its Na⁺ storage mechanism, particularly on the low potential plateau, remains controversial. To overcome this issue, two types of hard carbons with different microstructures are employed and the relationship between the microstructures and Na⁺ storage behaviors is evaluated. By the combination of operando X■ray diffraction, ex situ Raman spectroscopy, NMR, and theoretical calculation, it is found that the sodium storage capacities of the hard carbons in the low potential plateau region contain the concurrent contributions from both interlayer intercalation and micropores filling, and the ratio of the two contributors greatly depends on the microstructure of hard carbon materials. Moreover, an electrochemical pointer (potential inflection point at the end of the discharge curve) is found to distinguish the dominance of interlayer intercalation and the micropores filling processes of sodium ions in the low potential plateau region. Based on this new finding, a microstructure■dependent mechanism (“adsorption■intercalation/filling” hybrid mechanism) is proposed to achieve an overall understanding of the sodium storage behaviors in different hard carbon materials, which may provide deep insight into the rational design of hard carbon structures as high■performance anode materials for advanced SIBs.